Chin. Phys. Lett. **39**, 027301 (2022) **Express Letter**

A Theory for Anisotropic Magnetoresistance in Materials with Two Vector Order Parameters

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(Received 14 December 2021; accepted 30 December 2021; published online 6 January 2022)

Anisotropic magnetoresistance (AMR) and related planar Hall resistance (PHR) are ubiquitous phenomena of magnetic materials. Although the universal angular dependences of AMR and PHR in magnetic polycrystalline materials with one order parameter are well known, no similar universal relation for other class of magnetic materials are known to date. Here a general theory of galvanomagnetic effects in magnetic materials is presented with two vector order parameters, such as magnetic single crystals with a dominated crystalline axis or polycrystalline non-collinear ferrimagnetic materials. It is shown that AMR and PHR have a universal angular dependence. In general, both longitudinal and transverse resistivity are non-reciprocal in the absence of inversion symmetry: Resistivity takes different values when the current is reversed. Different from simple magnetic polycrystalline materials where AMR and PHR have the same magnitude, and $\pi/4$ out of phase, the magnitudes of AMR and PHR of materials with two vector order parameters are not the same in general, and the phase difference is not $\pi/4$. Instead of π periodicity of the usual AMR and PHR, the periodicities of materials with two order parameters are 2π.

DOI: 10.1088/0256-307X/39/2/027301

Anisotropic magnetoresistance (AMR) of magnetic materials has been a well-known phenomenon since Lord Kelvin in 1856,[1] then known as William Thomson, found that electrical resistances of a piece of nickel and iron are different when a current is parallel and perpendicular to the magnetization. This phenomenon and related planar Hall resistance (PHR) are technologically important in magnetic sensors and data storage and retrieval.[2,3] AMR and PHR have constantly attracted much attention with continuously improved understanding since their discoveries.[4−14] Phenomenologically and logically, spin-orbit interaction and s–d scatterings must play essential roles in the AMR, PHR, and extraordinary galvanomagnetic effects[2,3] in general because moving electrons "see" the magnetization (spins). Unfortunately, the exact origins of AMR and PHR are unclear to date despite of those progress.

AMR can, in principle, occur in all magnetic materials, but is more notable for good conducting materials such that magnetization dependent band structure and spin dependent scatterings contribute significantly to their resistances. It is typically a few per cent for metallic magnetic materials such as iron and nickel, and much less for amorphous materials whose resistances are mainly from other spin-independent scattering. However, there are also reports that AMR could be more than 50% in some single crystals.[11] To date, the only known universal behavior of AMR is its angular dependence in magnetic polycrystals when the only order parameter is the total magnetization. The anisotropic resistivity (or resistance) $\Delta\rho(\alpha)$ follows $\Delta\rho(\alpha) = \Delta\rho_0 \cos^2\alpha$, where α is the angle between the magnetization and current. The widely known theory for this universal law is the so-called two-current model proposed by Campbell, Fert and Jaoul.[2−4] The theory and its extensions[5,6] assume that current can be divided into the contributions from spin-up and spin-down electrons. Without s–d scatterings, two currents are independent of each other, magnetoresistance is determined by the shunted current. In the presence of s–d scatterings, two currents are partially mixed and the lift of shunting effect depends on the angle α between the magnetization and current. The theory leads to an approximate $\cos^2\alpha$-formula for AMR under certain assumptions and limits, in contrast to the exact $\cos^2\alpha$-law observed in many polycrystalline magnets. Furthermore, these theories require the exact angular distribution of atomic d-orbits for explaining the $\cos^2\alpha$-law, in contrast to the fact that d-electron wavefunctions in a polycrystal must deviate from their atomic counterparts at sub-nanometer scale due to the crystal fields. In one word, these theories cannot account for the exact universal angular dependences of AMR observed in many polycrystalline magnets originated from both d-electrons and f-electrons. Of course, it should be pointed out that much less-known theories based on symmetry

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argument[15] and tensor analysis[16] can indeed explain the universal AMR and PHR for polycrystalline magnetic materials. For the AMR and PHR in single crystals, there are also many studies[17–24] that show complicate behaves with limited understanding.[12,13] The observed behaviors cannot be explained by the two-current model[5,6] or those[15,16] for polycrystalline materials.

In this Letter, we derive a generic formula for extraordinary galvanomagnetic effects in magnetic materials with two vector order parameters. Materials could be polycrystalline ferrimagnets with two sublattices, or magnetic single crystals whose electron transport is dominated by one direction while the other two directions are the same. It could also be the helimagnets in helical states.[25] Our theory is based on the general requirement that all physical quantities must be tensors[26] and the laws of physics must be in tensor forms. Under this requirement, the AMR and PHR have universal forms in such magnetic materials that are distinct from the well-known AMR and PHR behaviors in simple polycrystalline magnetic materials with only one order parameter. The magnitude of AMR and PHR are not the same in general, and the phase difference is not $\pi/4$. Instead of π periodicity in the usual AMR and PHR, the periodicities of the AMR and PHR in the new class of materials are 2π.

Consider an infinite ferromagnetic single crystal with only one dominate crystalline axis $\boldsymbol{n}$ and a magnetization $\boldsymbol{M}$, which is not along $\boldsymbol{n}$. In the linear response region, the electric field $\boldsymbol{E}$ in response to an applied current density $\boldsymbol{J}$ in the crystal must be

$$\boldsymbol{E} = \rho(\boldsymbol{M}, \boldsymbol{n})\boldsymbol{J}, \tag{1}$$

where $\rho(\boldsymbol{M}, \boldsymbol{n})$ is a Cartesian tensor of rank 2. Although the tensor values depend on microscopic properties of the crystal and parameters that define the thermodynamic state, the tensor form can only come from the order parameters that characterize the macroscopic state of the system. In the absence of an external magnetic field, $\boldsymbol{M}$ and $\boldsymbol{n}$ are the only available vectors that can be used to construct tensor ρ. Thus, ρ should be the linear combination of $\boldsymbol{MM}$, $\boldsymbol{Mn}$, and $\boldsymbol{nn}$. Each of the three Cartesian tensors is not irreducible,[26] and can be decomposed into the direct sum of a scalar, a vector, and a traceless symmetric tensor. Thus, from $\boldsymbol{M}$ and $\boldsymbol{n}$, it is possible to construct three vectors and three traceless symmetric tensors of ranks 2. They are $\boldsymbol{M}$, $\boldsymbol{n}$, $\boldsymbol{M}\times\boldsymbol{n}$, $\boldsymbol{MM}-M^2/3$, $\boldsymbol{Mn}+\boldsymbol{nM}-2\boldsymbol{M}\cdot\boldsymbol{n}/3$, and $\boldsymbol{nn}-1/3$, where M is the magnitude of magnetization $\boldsymbol{M}$. Thus, with these six angular dependent terms together with a scaler term, the electric field $\boldsymbol{E}$ induced by $\boldsymbol{J}$, after grouping similar terms, must take the following most generic form

$$\begin{aligned}\boldsymbol{E} = {}& \rho_1\boldsymbol{J} + \boldsymbol{J}\times(R_1\boldsymbol{M} + R_2\boldsymbol{n}) + A_1(\boldsymbol{J}\cdot\boldsymbol{M})\boldsymbol{M} \\ & + A_2(\boldsymbol{J}\cdot\boldsymbol{n})\boldsymbol{n} + A_3\boldsymbol{J}\times(\boldsymbol{M}\times\boldsymbol{n}) + A_4(\boldsymbol{M}\cdot\boldsymbol{n})\boldsymbol{J},\end{aligned} \tag{2}$$

where $\rho_1 R_1$, R_2, A_i $(i = 1, 2, 3, 4)$ are parameters that are determined by the intrinsic and extrinsic properties of the sample such as the band structures and impurity specifics. Of course, these parameters can, in principle, depend on the scalers constructed from $\boldsymbol{M}$ and $\boldsymbol{n}$, such as M^2 and $\boldsymbol{M}\cdot\boldsymbol{n}$. Among them, only $\boldsymbol{M}\cdot\boldsymbol{n}$ can introduce the anisotropic effect. Since magnetic interaction are very weak, in terms of perturbation, only low powers of M^2 and $\boldsymbol{M}\cdot\boldsymbol{n}$ contribute mainly to these parameters. These terms depend on the relative directions among $\boldsymbol{J}$, $\boldsymbol{M}$, and $\boldsymbol{n}$. It may be worthwhile to point out that this is in the same sprite as those in the thermodynamics: The behavior of a given system can be uniquely determined by a few parameters. In the current case, $\boldsymbol{E}$ is determined by $\boldsymbol{J}$, $\boldsymbol{M}$, and $\boldsymbol{n}$ when other external parameters such as temperature and pressure do not vary.

In the case of polycrystalline sample, $\boldsymbol{n}$ is absent, and Eq. (2) reduces to the well known generalized Ohm's law of polycrystalline materials[16,27] with only ρ_1, R_1 and A_1 terms. R_1-term and A_1-term are the usual anomalous Hall effect and AMR and PHR, respectively. The longitudinal and transverse resistivity are $\rho_{xx} = \rho_1 + A_1M^2\cos^2\alpha$ and $\rho_{xy} = R_1M_z + \frac{A_1M^2}{2}\sin(2\alpha)$, if the $\hat{x}$ direction is defined along $\boldsymbol{J}$ and $\boldsymbol{M}$ is in the xy-plane with angle α between $\boldsymbol{M}$ and $\boldsymbol{J}$. Obviously, ρ_1 is the longitudinal resistivity when $\boldsymbol{J}$ is perpendicular to $\boldsymbol{M}$ and R_1 is the anomalous Hall coefficient. A_1M^2 is the amplitude of the conventional AMR and PHR that is typically a few percent of ρ_1.

It may be illustrative to consider two special setups of $\boldsymbol{J}$ parallel or perpendicular to $\boldsymbol{n}$. For $\boldsymbol{J}\parallel\boldsymbol{n}\parallel\hat{x}$, $\rho_{xx} \equiv \boldsymbol{E}\cdot\hat{x}/J = (\rho_1 + A_2) + A_1M^2\cos^2\alpha + A_4M\cos\alpha$ and $\rho_{xy} \equiv \boldsymbol{E}\cdot\hat{y}/J = R_1M_z + \frac{A_1M^2}{2}\sin(2\alpha) + A_3M\sin\alpha$. Interestingly and strangely, it predicts a non-reciprocal dc electron transport if the system is not invariant under the $\boldsymbol{n}\to-\boldsymbol{n}$ transformation. Namely, ρ_{xx} and ρ_{xy} take different values when current $\boldsymbol{J}$ is reversed. Phases of AMR and PHR do not differ by $\pi/4$, and their amplitudes are not the same. If the system is invariant under the $\boldsymbol{n}\to-\boldsymbol{n}$ transformation, then A_3 and A_4 must be odd functions of $\boldsymbol{M}\cdot\boldsymbol{n} = M\cos\alpha$. Assume that only leading orders of $A_3 = C_3M\cos\alpha$ and $A_4 = C_4M\cos\alpha$ exist, then $\rho_{xx} = (\rho_1 + A_2) + (A_1 + C_4)M^2\cos^2\alpha$ and $\rho_{xy} = R_1M_z + \frac{A_1+C_3}{2}M^2\sin(2\alpha)$. AMR and PHR follow the conventional $\cos^2\alpha$ and $\sin(2\alpha)$ laws and have the same phase lag but with different amplitudes, in general.

For $\boldsymbol{J}\parallel\hat{x}\perp\boldsymbol{n}\parallel\hat{z}$, AMR behaves differently for $\boldsymbol{M}$

varying in the xy- and yz-, and zx-planes. Here $\rho_{xx} = \rho_1 + A_1 M^2 \cos^2\alpha$ for $\boldsymbol{M}$ in the xy-plane with α being the angle between $\boldsymbol{M}$ and $\boldsymbol{J}$, $\rho_{xx} = \rho_1 + A_4 M \cos\beta$ for $\boldsymbol{M}$ in the yz-plane with β being the angle between $\boldsymbol{M}$ and $\boldsymbol{n}$, and $\rho_{xx} = \rho_1 + A_1 M^2 \sin^2\gamma + A_4 M \cos\gamma$ for $\boldsymbol{M}$ in the zx-plane with γ being the angle between $\boldsymbol{M}$ and $\boldsymbol{n}$. With the $\boldsymbol{n} \to -\boldsymbol{n}$ symmetry and with the same reason mentioned above, we have $\rho_{xx}(\alpha) = \rho_1 + A_1 M^2 \cos^2\alpha$, $\rho_{xx}(\beta) = \rho_1 + C_4 M^2 \cos^2\beta$, and $\rho_{xx}(\gamma) = \rho_1 + A_1 M^2 + (C_4 - A_1) M^2 \cos^2\gamma$. Clearly, this is very different from the conventional AMR that does not have angular dependence for $\boldsymbol{M}$ in the yz-plane.

Magnetic materials do not respect the time-reversal symmetry because a current reverses its direction under the transformation, and so does the related magnetization. Both spins and magnetization do not change under an inversion transformation, whereas a current reverses its direction under the transformation. Thus, magnetic materials without inversion symmetry are good candidates for observing the non-reciprocal dc electronic transport. This can happen for chiral magnets with the Dzyaloshinskii–Moriya interactions (DMI). Like the ubiquity of spin-orbit interactions for all materials, DMI universally exists in all magnetic materials. Thus, their resistivity should be non-reciprocal in principle, and the issue is only whether the non-reciprocity is strong enough to be measurable. In the case that Eq. (2) is invariant under the inversion, $\boldsymbol{x} \to -\boldsymbol{x}$, ρ_1, R_1, A_1, and A_2 must be even functions of $\boldsymbol{M}\cdot\boldsymbol{n}$, and R_2, A_3, and A_4 are odd since both $\boldsymbol{E}$ and $\boldsymbol{J}$ change sign. Thus, the non-reciprocity of resistivity disappears in a system with the inversion symmetry, and the angular dependence of AMR and PHR is the same as those for the polycrystalline magnetic material with only one order parameter. For polycrystalline magnetic-film/heavy-metal bilayer such as Co/Pt, the system has the in-plane inversion symmetry, but without mirror symmetry with respect to the interface. In this case, one expects reciprocity of the longitudinal resistivity in the plane and non-reciprocity in tunnelling resistance. Of course, one should anticipate difficulty in detecting the interfacial effect in metallic systems.

The approach stated above may offer a natural explanation to the strange angular dependences of ρ_{xx} in body-centered cubic (bcc) CoFe single crystal when $\boldsymbol{J} \parallel \hat{x}$ is along the magnetic crystalline easy-axis [1,1,0] ($\boldsymbol{n}_1$) and the film deposited direction is along [0,0,1] (the $\hat{z}$-direction) and the $\hat{y}$ axis is along another equivalent easy-axis of $[1,\bar{1},0]$ ($\boldsymbol{n}_2$).[12] The distinct features include identical strong two-fold AMR when $\boldsymbol{M}$ varies in the yz- and zx-planes, and a weak four-fold AMR when $\boldsymbol{M}$ varies in the xy-plane. Although CoFe is not a single crystal with only one axis, one can still construct relevant terms in $\boldsymbol{E}$ by treating $\boldsymbol{n}_1$ and $\boldsymbol{n}_2$ as equivalent vectors. Then $\boldsymbol{E} = \rho_1\boldsymbol{J} + \boldsymbol{J}\times R_1\boldsymbol{M} + A_1 J M_x^2 + A_2 J\boldsymbol{n}_1 + A_4(\boldsymbol{M}\cdot\boldsymbol{n}_1)\boldsymbol{J} + A_4'(\boldsymbol{M}\cdot\boldsymbol{n}_2)\boldsymbol{J}$, R_2- and A_3-terms in Eq. (2) are absent because the equivalent vectors $\pm\boldsymbol{n}_i$ ($i = 1,2$) cancel each other. A_1 must be even in $\boldsymbol{M}\cdot\boldsymbol{n}_1$ and $\boldsymbol{M}\cdot\boldsymbol{n}_2$, and A_4 and A_4' must be odd because $\boldsymbol{E}$ should be symmetric under $\boldsymbol{n}_i \to -\boldsymbol{n}_i$ (i = 1,2) transformations for bcc CoFe. If we take $A_1 = C_1(\boldsymbol{M}\cdot\boldsymbol{n}_2)^2$, $A_4 = C_4\boldsymbol{M}\cdot\boldsymbol{n}_1$ and $A_4' = C_4\boldsymbol{M}\cdot\boldsymbol{n}_2$, the longitudinal resistivity will become $\rho_{xx} = \rho_1 + C_1 J M_x^2 M_y^2 + C_4 M_x^2 + C_4 M_y^2$. It gives $\rho_{xx}(\alpha) = \rho_1' + C_1 J M^4 \cos 4\alpha$, $\rho_{xx}(\beta) = \rho_1 + C_4 M^2 \sin^2\beta$, and $\rho_{xx}(\gamma) = \rho_1 + C_4 M^2 \sin^2\gamma$, where α is the angle between $\boldsymbol{M}$ and $\boldsymbol{J}(\hat{x})$ when $\boldsymbol{M}$ varies in the xy-plane; β and γ are the angles between $\boldsymbol{M}$ and $\hat{z}$ when $\boldsymbol{M}$ varies in the yz- and zx-planes, respectively. Since C_1 is the 4th order (M^4) and C_4 is the second order (M^2) in perturbation, one expects $C_4 \gg C_1$. Thus, four-fold AMR of $\rho_{xx}(\alpha)$ is much weaker than the two identical two-fold AMR of $\rho_{xx}(\beta) = \rho_{xx}(\gamma)$. This is very unusual because the conventional AMR has a strong two-fold $\rho_{xx}(\alpha)$, and no or insignificant angular dependence of $\rho_{xx}(\beta)$ because $\boldsymbol{J} \perp \boldsymbol{M}$. These unusual angular dependences of ρ_{xx} on orientation of $\boldsymbol{M}$ is exactly what was observed in the recent experiment.[12] If C_1 and C_4 are the intrinsic mechanisms of AMR that come from the modification of band by magnetization $\boldsymbol{M}$ when it aligns along the crystalline axis [1,1,0] and $[1,\bar{1},0]$, then the above $\boldsymbol{E}$ expression makes a lot of sense. It may be important to emphasize that the above analysis works only when the current is along $[1,1,0]$. Furthermore, the above analysis depends not only on bcc CoFe, but also on the assumption that, somehow, only two equivalent directions of $[1,1,0]$ and $[1,\bar{1},0]$, but not others, are important. The analysis provides only a possible understanding of the recent surprising experimental observations, and more study is needed.

Magnetic single crystals with only one dominated axis are not very common because a crystal has three principle axis by definition although many tetrahedron and hexagonal structures are believed to be uniaxial magnets. To test the theory presented above, it may be useful to find easily realizable materials where the generalization of the theory applies. Real crystals may have other rotational and mirror symmetries that provide extra macroscopic orders. There would be many more terms in Eq. (2) and above simple universal behavior disappear. Thus, more realistic materials to test the theory may be polycrystalline materials. As clearly shown in our derivation, the theory is based on the assumption of two vector order parameters. Thus, the theory is applicable to non-collinear ferrimagnetic polycrystals with two sub-latices. The magnetizations $\boldsymbol{M}_1$ and $\boldsymbol{M}_2$ of the two sub-lattices are the only available order parameters, and we need only to replace $\boldsymbol{M}$ and $\boldsymbol{n}$ in Eq. (2) by $\boldsymbol{M}_1$ and $\boldsymbol{M}_2$. The resulted

equation is

$$\begin{aligned}\boldsymbol{E} = {} & \rho_1 \boldsymbol{J} + \boldsymbol{J} \times (R_1 \boldsymbol{M}_1 + R_2 \boldsymbol{M}_2) + A_1 (\boldsymbol{J} \cdot \boldsymbol{M}_1) \boldsymbol{M}_1 \\ & + A_2 (\boldsymbol{J} \cdot \boldsymbol{M}_2) \boldsymbol{M}_2 + A_3 \boldsymbol{J} \times (\boldsymbol{M}_1 \times \boldsymbol{M}_2) \\ & + A_4 (\boldsymbol{M}_1 \cdot \boldsymbol{M}_2) \boldsymbol{J}. \end{aligned} \tag{3}$$

Of course, $|\boldsymbol{M}_2| = M_2 \neq 1$ now.

Consider a special case where $\boldsymbol{J}$, $\boldsymbol{M}_1$, and $\boldsymbol{M}_2$ are in a plane with the angles between $\boldsymbol{J}$ and $\boldsymbol{M}_1$, $\boldsymbol{M}_2$ being θ and ϕ. The longitudinal and transverse resistivities read

$$\begin{aligned} \rho_{xx} &= \rho_1 + \rho_2 \cos^2\theta + \rho_3 \cos^2\phi + \rho_4 \cos(\phi - \theta), \\ \rho_{xy} &= \rho_5 + \frac{\rho_2}{2} \sin(2\theta) + \frac{\rho_3}{2} \sin(2\phi) + \rho_6 \sin(\phi - \theta), \end{aligned} \tag{4}$$

where $\rho_2 \equiv A_1 M_1^2$, $\rho_3 \equiv A_2 M_2^2$, $\rho_4 \equiv A_4 M_1 M_2$, $\rho_5 \equiv R_1 M_{1z} + R_2 M_{2z}$, and $\rho_6 \equiv A_3 M_1 M_2$. When $\phi - \theta = 180°$, or for collinear ferrimagnetic crystalline, Eq. (4) returns to the well-known AMR and PHR. It is well known that a collinear antiferromagnet undergoes a spin-flop transition when a magnetic field along the Neel order parameter is larger than a critical value. Thus, the current theory may be best tested in a collinear antiferromagnet. The fingerprint is the change of angular dependences of AMR and PHR before and after spin-flop transition: Before the transition, AMR and PHR have the same amplitudes and follow the $\cos^2\theta$ law and the $\sin(2\theta)$ law, respectively. After the transition, AMR and PHR are described by Eq. (4). If the antiferromagnet has a strong DMI such that the system does not have inversion symmetry, the magnitudes of AMR and PHR of the materials with two vector order parameters are not the same, as discussed earlier, and the phase difference is not $\pi/4$. Instead of π periodicity of the AMR and PHR in polycrystalline materials with only one order parameter, the periodicities of the AMR and PHR in Eq. (4) are 2π.

Our predictions are based on the tensor forms of the laws of physics. This is the same approach for Einstein's gravitational law. The argument is that the Ricci tensor and metric tensor are the only possible tensors of rank 2 out of the metric tensor while Newtonian gravitation law was identified as an equation between one metric tensor component and the energy.[28] Thus, a proper linear combination of Ricci tensor and Ricci curvature (scaler) multiplying metric tensor is equal to the energy-momentum tensor. The method was also used in condensed matter physics for Ohm's law in ferromagnet[27] and for the anomalous spin-Hall effects (ASHEs) and anomalous inverse spin-Hall effects (AISHEs),[29,30] which is also called magnetization dependent spin-Hall and inverse spin-Hall effects. Interestingly, our analysis predicts that $\rho_{xx}(\theta = 180°) \neq \rho_{xx}(\theta = 0°)$ in general. However, $\rho_{xx}(\theta = 180°) = \rho_{xx}(\theta = 0°)$ holds in a material if it has the inversion symmetry. This is because of $\boldsymbol{E}(-\boldsymbol{J}) = -\boldsymbol{E}(\boldsymbol{J})$ under the symmetry. The AMR and PHR in such a material behave the same as those in a polycrystal with only one order parameter.

It should be pointed out that irreducible tensor decomposition in our derivation of Eq. (2) is important because different irreducible tensors vary with other parameters independently although they are from the same Cartesian tensors. For systems with two vector order parameters, ρ_1, $R_i (i = 1, 2)$, and $A_i (i = 1, 2, 3, 4)$ are the seven independent material parameters in the galvanomagnetic effects. The general expressions of Eqs. (2) and (3) do not distinguish an intrinsic mechanism such as the band structure contribution from an extrinsic mechanism such as spin-dependent electron scattering due to defects and phonons. Our predictions were not derived from a microscopic Hamiltonian, and the theory does not provide actual values of the seven parameters. In principle, their values can be computed from a given microscopic model using quantum mechanics. Such a microscopic theory is surely important and necessary although it is foreseeable to be not easy because of too many process in real materials.

In summary, a generic galvanomagnetic effect in magnetic materials whose thermodynamic states can be described by two vector order parameters. It is found that the AMR and PHR in such materials have universal angular dependences. In chiral magnets, for example, materials with DMI, both longitudinal and transverse dc resistivities are predicted to be nonreciprocal. Different from polycrystalline magnetic materials with only one order parameter where AMR and PHR have the same magnitude, and $\pi/4$ out of phase, the magnitude of AMR and PHR of materials with two order parameters are not the same in general, and the phase difference is not $\pi/4$. Instead of π periodicity of the usual AMR and PHR, the periodicities of AMR and PHR in magnetic materials with two order parameters are 2π.

Acknowledgments. This work was supported by the National Key Research and Development Program of China (Grant Nos. 2020YFA0309600 and 2018YFB0407600), the National Natural Science Foundation of China (Grant Nos. 11974296 and 11774296), and Hong Kong RGC (Grant Nos. 16301518, 16301619, and 16302321).

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